Line width of magnetic resonance exchange modes in a four-sublattice orthorhombic antiferromagnet

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The contributions of magnon-magnon and magnon-phonon interaction processes to the line width of magnetic resonance exchange modes in a four-sublattice orthorhombic antiferromagnet have been investigated. A comparison is made with experimental results for $CuCl_2 \cdot 2H_2O$. In this crystal the main contribution to relaxation is determined by decay processes, and decay processes governed by magnetoelastic interactions of exchange origin are dominant.

1. INTRODUCTION

Most known magnetically ordered crystals are multisublattice magnets. Investigations of those specific characteristics of such magnets that depend on the presence of a large number of magnetic sublattices were started comparatively recently. Attention was focused primarily on the spectrum of elementary excitations of the magnetic subsystem. The upper branches of the magnetic resonance spectrum, the so-called exchange modes,[1–6] were investigated. These branches of the spectrum, which are the analog of optical phonons, are interesting because their activation energies are determined in the exchange approximation by intersublattice exchange integrals and the corresponding frequencies for the three-dimensional magnets fall into the submillimeter wavelength range.

The frequency-field dependence of the magnetic resonance spectrum of a series of multisublattice magnets, the characteristics of the exchange–spin-wave spectrum, and the associated specific character, of two-magnon absorption on the exchange branches of the spectrum, as well as Raman scattering of light by magnons in multisublattice magnets have now been studied in detail.[1–6]

The usual problem addressed in investigations of the dynamic properties of magnets is the study of their relaxational characteristics—the widths of the magnetic-resonance lines and the damping coefficients of the spin waves. Until now the relaxational characteristics of multisublattice magnets have been studied only on the basis of a phenomenological approach[7–9] in Refs. 10 and 11. The microscopic relaxation mechanisms have been analyzed in detail only for ferromagnets and for two-sublattice antiferromagnets,[12–14] i.e., magnon and magnon-phonon interaction processes have been investigated only for acoustic magnons.

The purpose of this work is to study systematically the microscopic mechanisms of relaxation of exchange spin waves in a four-sublattice orthorhombic antiferromagnet. The magnon-magnon and magnon-phonon interaction processes are analyzed, the contributions of these processes to the line width of the magnetic resonance exchange modes are calculated, and the computational results are compared with experimental data for the antiferromagnet $CuCl_2 \cdot 2H_2O$.

The exchange-magnon relaxation processes studied in this work can be conventionally divided into three groups. These processes are governed by the magnon interactions within the exchange-magnon subsystem, the interaction of exchange and acoustic magnons, and by processes describing the interaction of the magnon and phonon subsystems. In the exchange-magnon subsystem the laws of conservation of energy and momentum allow only scattering of the exchange magnons by one another. In the interaction of exchange and acoustic magnons, aside from scattering processes, decay processes (for example, the decay of an exchange magnon into three acoustic magnons) are also possible. A specific feature of decay processes is that their amplitudes are determined by the nonuniform Dzyaloshinskiĭ–Moriya interaction. The types of allowed interactions of exchange magnons and phonons depend, however, on the relation between the Debye temperature and the exchange integrals. Thus for the example of the four-sublattice antiferromagnet $CuCl_2 \cdot 2H_2O$, which we studied, the Debye temperature is greater than all characteristic intersublattice exchange integrals, and the process in which an exchange magnon decays into an acoustic magnon and a phonon is allowed. The amplitude of this process is proportional to the exchange magnetoelastic constant. As will be shown below, decay processes play the dominant role in the relaxation of exchange modes.

2. STRUCTURE OF THE HAMILTONIAN

We represent the Hamiltonian of the magnet in the form

$$\hat{H}=\hat{H}_m+\hat{H}_{mp}+\hat{H}_p, \tag{1}$$

where $\hat{H}_m$, $\hat{H}_{mp}$, and $\hat{H}_p$ describe the magnetic subsystem, the magnetoelastic interactions, and the phonon subsystem, respectively.

From the viewpoint of symmetry the type of antiferromagnet in which we are interested corresponds to the situation when the components of the principal vectors of antiferromagnetism and magnetization transform according to different irreducible representations of the symmetry group of the paramagnetic phase. The existence of the Dzyaloshinskiĭ-Moriya interaction in this case can result in "bending" of the spins of the sublattices and the appearance of components of weak-antiferromagnetism vectors.[15] In or-

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der to take into account as much as possible the symmetry of the magnet we introduce the linear combinations of the Fourier components $s_\alpha(\mathbf{k})$ of the spins of the sublattices, where α is the sublattice index and $\mathbf{k}$ is the wave vector:

$$\begin{aligned} \mathbf{L}_1(\mathbf{k})&=\mathbf{s}_1(\mathbf{k})+\mathbf{s}_2(\mathbf{k})-\mathbf{s}_3(\mathbf{k})-\mathbf{s}_4(\mathbf{k}),\\ \mathbf{L}_2(\mathbf{k})&=\mathbf{s}_1(\mathbf{k})-\mathbf{s}_2(\mathbf{k})+\mathbf{s}_3(\mathbf{k})-\mathbf{s}_4(\mathbf{k}),\\ \mathbf{L}_3(\mathbf{k})&=\mathbf{s}_1(\mathbf{k})-\mathbf{s}_2(\mathbf{k})-\mathbf{s}_3(\mathbf{k})+\mathbf{s}_4(\mathbf{k}),\\ \mathbf{F}(\mathbf{k})&=\mathbf{s}_1(\mathbf{k})+\mathbf{s}_2(\mathbf{k})+\mathbf{s}_3(\mathbf{k})+\mathbf{s}_4(\mathbf{k}). \end{aligned} \tag{2}$$

For $\mathbf{k}=0$ the components of these vectors are the basis functions of the irreducible representation of the symmetry group D_{2h}^{7} of the paramagnetic phase of $CuCl_2\cdot 2H_2O$. The classification of these basis functions is presented in Ref. 1. The general form of the Hamiltonian of the magnetic subsystem can be written as follows:

$$\hat{H}_m=\sum \mathscr{K}_{\alpha\beta}{}^{ij}(\mathbf{k})s_\alpha{}^i(-\mathbf{k})s_\beta{}^j(\mathbf{k})-\mu_B N^{1/2}\sum g_\alpha{}^{ij}H_i s_\alpha{}^j(0), \tag{3}$$

where $\mathscr{K}_{\alpha\beta}^{ij}(\mathbf{k})$ describes the isotropic and antisymmetric exchange interactions as well as the relativistic interactions between the sublattices α and β; the indices i and j refer to the Cartesian coordinates; N is the number of magnetic cells in the crystal; μ_B is the Bohr magneton; and, g_α^{ij} is the g-factor tensor of the αth ion. Using Eq. (2), we can put the expression (3) into the form

$$\begin{aligned} \hat{H}_m=\sum_{\mathbf{k}}\Big\{\sum_i\Big[&J_{0i}(\mathbf{k})F_i(-\mathbf{k})F_i(\mathbf{k})\\ &+\sum_\sigma J_{\sigma i}(\mathbf{k})L_{\sigma i}(-\mathbf{k})L_{\sigma i}(\mathbf{k})\Big]\\ &+D_{01}(\mathbf{k})F_x(-\mathbf{k})L_{2z}(\mathbf{k})+D_{02}(\mathbf{k})F_z(-\mathbf{k})L_{2x}(\mathbf{k})\\ &+D_{03}(\mathbf{k})L_{1x}(-\mathbf{k})L_{3z}(\mathbf{k})\\ &+D_{04}(\mathbf{k})L_{1z}(-\mathbf{k})L_{3x}(\mathbf{k})\\ &-N^{1/2}\Delta(\mathbf{k})[g_1H_x(F_x(\mathbf{k})+\tau_1L_{2z}(\mathbf{k}))+g_2H_yF_y(\mathbf{k})\\ &+g_3H_z(F_z(\mathbf{k})+\tau_3L_{2x}(\mathbf{k}))]+\hat{H}_1(\mathbf{k})\Big\}. \end{aligned} \tag{4}$$

Here the quantities J and D are linear combinations of the exchange intersublattice integrals of the type (2) (see Ref. 1). In the standard three-dimensional antiferromagnets the relations

$$J_{\sigma i}\gg D\gg|J_{\sigma i}-J_{\sigma j}|,\quad i\neq j, \tag{5}$$

are satisfied,[15] since D is determined by the Dzyaloshinskiĭ-Moriya interaction and the quantity $|J_{\sigma i}-J_{\sigma j}|\approx A$ is the anisotropy. The Hamiltonian $\hat{H}_1(\mathbf{k})$ contains terms which are determined by the exchange-relativistic and relativistic interactions, which vanish for $\mathbf{k}=0$. In each specific case the form of $\hat{H}_1(\mathbf{k})$ can be obtained starting from the characteristics of the crystal and magnetic structures of the magnet. In what follows we shall include in $\hat{H}_1(\mathbf{k})$ only the terms arising owing to the nonuniform Dzyaloshinskiĭ interaction. Such terms were studied previously in Ref. 16 in an investigation of the high-frequency properties of yttrium iron garnet.

We shall examine the mechanism responsible for their appearance. The Dzyaloshinskiĭ–Moriya interaction is described by the off-diagonal components of the quantities

$$\mathscr{K}_{\alpha\beta}{}^{ij}(\mathbf{k})=N^{-1}\sum_n \mathscr{K}_{\alpha\beta}{}^{ij}(n,m)\exp(i\mathbf{k}\mathbf{r}_{nm}), \tag{6}$$

where n and m are the numbers of the unit cells. According to Ref. 15, the symmetric part of the off-diagonal components is of the order of $(\Delta g/g)^2\mathscr{K}_{\alpha\beta}^{ij}$ while the antisymmetric part is of the order of $(\Delta g/g)\mathscr{K}_{\alpha\beta}^{ij}$, where Δg is the deviation of the g-factor from its free-electron value.

The positional symmetry of the pair of sublattices α and β makes it possible to determine the zero components of the quantities $\mathscr{K}_{\alpha\beta}^{ij}(0)$ and to indicate which nonzero components are purely symmetric, purely antisymmetric, or mixed. In so doing, symmetry operations which do not permute the sublattices as well as the relation $\mathscr{K}_{\alpha\beta}^{ij}(0)=\mathscr{K}_{\beta\alpha}^{ji}(0)$ are employed. It is obvious that the symmetry of the tensors $\mathscr{K}_{\alpha\beta}^{ij}(n,m)$ is lower than that of $\mathscr{K}_{\alpha\beta}^{ij}(0)$, since in order to investigate their forms it is necessary to use symmetry operations that do not affect the indices α and β as well as the numbers of the cells n and m. For this reason, the tensors $\mathscr{K}_{\alpha\beta}^{ij}(n,m)$ contain off-diagonal components, which are determined by the Dzyaloshinskiĭ–Moriya interaction and are not present in the quantities $\mathscr{K}_{\alpha\beta}^{ij}(0)$. After summing in Eq. (6) over cells with $\mathbf{k}=0$ these components cancel one another. For $\mathbf{k}\neq 0$, however, this cancellation does not occur and it is possible to talk about the terms of the Hamiltonian (3) that arise as a result of the nonuniform Dzyaloshinskiĭ–Moriya interaction.

In our case $\hat{H}_1(\mathbf{k})$ has the form

$$\begin{aligned}\hat{H}_1(\mathbf{k})=\tilde{R}(\mathbf{k})[&F_z(-\mathbf{k})L_{2y}(\mathbf{k})-F_y(-\mathbf{k})L_{2z}(\mathbf{k})\\ &+L_{1z}(-\mathbf{k})L_{3y}(\mathbf{k})-L_{1y}(-\mathbf{k})L_{3z}(\mathbf{k})].\end{aligned} \tag{7}$$

In deriving Eq. (7) we have retained the largest terms, corresponding to the interaction between the sublattices 1 and 2 as well as the sublattices 3 and 4. In the nearest-neighbor approximation the constant characterizing the nonuniform Dzyaloshinskiĭ interaction is determined by the expression

$$\tilde{R}(\mathbf{k})=2\mathscr{K}_{12}{}^{yz}\sin(\mathbf{ka}/2)\sin(\mathbf{kb}/2), \tag{8}$$

where $\mathbf{a}$ and $\mathbf{b}$ are lattice constants.

In what follows we confine our attention to the decay of exchange spin waves in a magnetic field oriented parallel to the easy axis of the crystal (x axis) and not exceeding the field in which the spin-flop transition occurs. In this case, in $CuCl_2\cdot 2H_2O$, aside from the main antiferromagnetism vector $L_{1x}\approx 4s$ and the weak-antiferromagnetism vector $L_{3z}\approx DL_{1x}/J$, the magnetic field gives rise to magnetization $F_x\approx HL_{1x}/J(D/J)^2$ and the additional weak-antiferromagnetism vector $L_{2z}\approx HDL_{1x}/J^2$ (Ref. 1).

Next, for the calculations we shall employ the method of second quantization developed in Ref. 5 for multisublattice magnets. The irreducible combinations of the operators (2) must be expressed in terms of the operators s'_α, each of which is written in its own local coordinate system with the z' axis directed along the equilibrium value of the spin. The transition matrices for different ions, which can be interchanged by symmetry operations, are related to one another.[5] This makes it possible to find a relation between the irreducible combinations $\mathbf{L}$ and the combinations $\mathbf{L}'$ of the form (2), written in the local coordinate system. In our case we have

$$\begin{pmatrix} F_x \\ L_{3y} \\ L_{2z} \end{pmatrix} = \hat{p} \begin{pmatrix} F_x' \\ F_y' \\ L_{1z}' \end{pmatrix} + \delta\hat{p} \begin{pmatrix} L_{1x}' \\ L_{1y}' \\ F_z' \end{pmatrix},$$

$$\begin{pmatrix} L_{1x} \\ L_{2y} \\ L_{3z} \end{pmatrix} = \hat{p} \begin{pmatrix} L_{1x}' \\ L_{1y}' \\ F_z' \end{pmatrix} + \delta\hat{p} \begin{pmatrix} F_x' \\ F_y' \\ L_{1z}' \end{pmatrix},$$

$$\begin{pmatrix} L_{3x} \\ F_y \\ L_{1z} \end{pmatrix} = \hat{p} \begin{pmatrix} L_{3x}' \\ L_{3y}' \\ L_{2z}' \end{pmatrix} + \delta\hat{p} \begin{pmatrix} L_{1x}' \\ L_{1y}' \\ L_{3z}' \end{pmatrix}, \tag{9}$$

$$\begin{pmatrix} L_{2x} \\ L_{1y} \\ F_z \end{pmatrix} = \hat{p} \begin{pmatrix} L_{1x}' \\ L_{1y}' \\ L_{3z}' \end{pmatrix} + \delta\hat{p} \begin{pmatrix} L_{3x}' \\ L_{3y}' \\ L_{2z}' \end{pmatrix},$$

where

$$\hat{p} = \begin{pmatrix} 0 & -a & -b \\ 1 & 0 & 0 \\ 0 & b & a \end{pmatrix}, \quad \delta\hat{p} = \begin{pmatrix} 0 & -c & -e \\ 0 & 0 & 0 \\ 0 & e & c \end{pmatrix}.$$

Here $a = L_{3z}/4s$, $b = L_{1x}/4s$, $c = L_{2z}/4s$, $e = F_x/4s$. The transformation from the operators L' to the spin-deflection operators is made with the help of the Goldstein-Primakov transformation:

$$F_x'(\mathbf{k}) = (\sigma s)^{1/2}\left[Q_F(k) - \frac{1}{4\sigma s N}\sum_{\mathbf{q},\mathbf{p}} V_T^+(\mathbf{q})\hat{B}_F(-\mathbf{p})V(\mathbf{k}+\mathbf{q}+\mathbf{p})\right]. \tag{10}$$

Here

$$V(\mathbf{k}) = \begin{pmatrix} a_F(\mathbf{k}) \\ a_{L_1}(\mathbf{k}) \\ a_{L_2}(\mathbf{k}) \\ a_{L_3}(\mathbf{k}) \end{pmatrix},$$

$$B_F(\mathbf{p}) = \begin{pmatrix} Q_F(\mathbf{p}) & Q_{L_1}(\mathbf{p}) & Q_{L_2}(\mathbf{p}) & Q_{L_3}(\mathbf{p}) \\ Q_{L_1}(\mathbf{p}) & Q_F(\mathbf{p}) & Q_{L_3}(\mathbf{p}) & Q_{L_2}(\mathbf{p}) \\ Q_{L_2}(\mathbf{p}) & Q_{L_3}(\mathbf{p}) & Q_F(\mathbf{p}) & Q_{L_1}(\mathbf{p}) \\ Q_{L_3}(\mathbf{p}) & Q_{L_2}(\mathbf{p}) & Q_{L_1}(\mathbf{p}) & Q_F(\mathbf{p}) \end{pmatrix}, \tag{11}$$

where

$$\begin{aligned} a_F(\mathbf{k}) &= {}^1/_2[a_1(\mathbf{k})+a_2(\mathbf{k})+a_3(\mathbf{k})+a_4(\mathbf{k})], \\ a_{L_1}(\mathbf{k}) &= {}^1/_2[a_1(\mathbf{k})+a_2(\mathbf{k})-a_3(\mathbf{k})-a_4(\mathbf{k})], \\ a_{L_2}(\mathbf{k}) &= {}^1/_2[a_1(\mathbf{k})-a_2(\mathbf{k})+a_3(\mathbf{k})-a_4(\mathbf{k})] \\ a_{L_3}(\mathbf{k}) &= {}^1/_2[a_1(\mathbf{k})-a_2(\mathbf{k})-a_3(\mathbf{k})+a_4(\mathbf{k})], \end{aligned} \tag{12}$$

$a_\alpha(\mathbf{k})$ is the operator of spin deflections of the α sublattice; σ is the number of sublattices; and,

$$Q_L(\mathbf{k}) = 2^{-1/2}[a_L^+(-\mathbf{k}) + a_L(\mathbf{k})].$$

The " + " sign in the vector $V(\mathbf{k})$ in the formula (11) indicates hermitian conjugation and transposition. An expression for L'_{1x} is obtained from Eq. (10) by changing the index in the first term $F\rightarrow L_1$ and in the matrix B_F by making the substitution $F\leftrightarrow L_1$ and $L_2\leftrightarrow L_3$. Analogously, for L'_{2x} in the first term of Eq. (10) it is necessary to make the substitution $F\rightarrow L_2$ and in the matrix B_F the substitutions $F\leftrightarrow L_2$ and $L_1\leftrightarrow L_3$, and also for L'_{3x} the substitution $F\rightarrow L_3$ in the first term of Eq. (10) and the substitutions $F\leftrightarrow L_3$ and $L_1\leftrightarrow L_2$ in the matrix B_F. The formulas for L'_y are obtained from Eqs. (10) and (11) by making the substitution

$$Q_L(\mathbf{k}) \rightarrow -iP_L(-\mathbf{k}) = -2^{-1/2}i[a_L^+(-\mathbf{k}) - a_L(\mathbf{k})].$$

For the z-components of the irreducible operators we have

$$\begin{aligned} F_z'(\mathbf{k}) &= 4sN^{1/2}\Delta(\mathbf{k}) + {}^1/_2N^{-1/2}\sum_{L\mathbf{q}\mathbf{p}}[P_L(\mathbf{q})P_L(-\mathbf{p}) - Q_L(\mathbf{q}) \\ &\quad\times Q_L(-\mathbf{p})]\Delta(\mathbf{q}+\mathbf{p}-\mathbf{k}), \\ L_{1z}'(\mathbf{k}) &= N^{-1/2}\sum_{\mathbf{q}\mathbf{p}}[P_F(-\mathbf{q})P_{L_1}(-\mathbf{p}) \\ &\quad+ P_{L_2}(-\mathbf{q})P_{L_3}(-\mathbf{p}) - Q_F(\mathbf{q})Q_{L_1}(-\mathbf{p}) \\ &\quad- Q_{L_2}(\mathbf{q})Q_{L_3}(-\mathbf{p})]\Delta(\mathbf{q}+\mathbf{p}-\mathbf{k}), \\ L_{2z}'(\mathbf{k}) &= L_{1z}'\{L_1\leftrightarrow L_2\}, \quad L_{3z}'(\mathbf{k}) \\ &= L_{1z}'\{L_1\leftrightarrow L_3\}. \end{aligned} \tag{13}$$

We substitute Eqs. (10) and (13) into the Hamiltonian (6) and drop all terms of order higher than fourth in the spin-deflection operators a_L. Terms of third order in these operators do not occur because of the symmetry of the given antiferromagnet. The quadratic part of the Hamiltonian obtained can be diagonalized with the help of the standard u-v transformation. In the absence of a magnetic field and the nonuniform Dzyaloshinskiĭ interaction the quadratic part of the Hamiltonian (6) gives a spin-wave spectrum consisting of two acoustic and two exchange magnon branches of different symmetry with energies ε_{47}, ε_{38} and ε_{16}, ε_{25}, respectively, where the subscripts enumerate the irreducible representations according to which the components of the vectors (2) participating in the oscillations on the given branch transform.

The magnetic field leads to "entanglement" of the spectra of the pair of exchange and pair of acoustic magnons;[3,4] as a result the components of the vectors (2) participating in the oscillations on this branch now transform according to four irreducible representations. Thus for two exchange branches we have $\varepsilon^{\pm}_{1256}$, while for the acoustic branches we have $\varepsilon^{\pm}_{3478}$. The form of the spin-wave spectrum of $CuCl_2 \cdot 2H_2O$ for $H = 4$ kOe is presented in Fig. 1. Taking into account the nonuniform Dzyaloshinskiĭ interaction results in complete mixing of the spin-wave states, if $k_x, k_y \neq 0$. In the absence of a magnetic field the part $\hat{H}_1(\mathbf{k})$ that is quadratic in the creation-annihilation operators mixes the states of exchange and acoustic magnons, ε_{16} and ε_{47}, respectively, as well as ε_{25} and ε_{38}; in addition, their interaction constant contains, aside from $R(\mathbf{k})$, an exchange-enhanced (at small $\mathbf{k}$) u-v coefficient of the acoustic magnon.

In the range of fields and wave vectors of interest to us the change in the energies of the spin waves governed by $\hat{H}_1(\mathbf{k})$ can nonetheless be neglected because of the smallness of the quantity R/J. The mixing of the states of the exchange

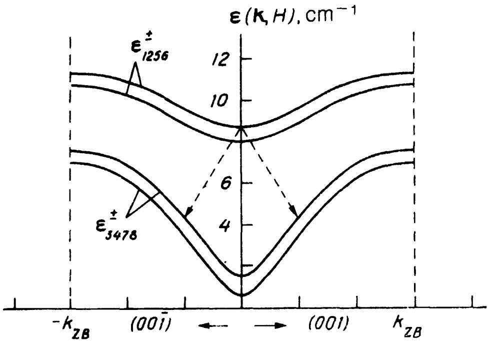

FIG. 1. The spin wave spectrum of $CuCl_2 \cdot 2H_2O$. The dashed arrows show one of the variants of the process, governed by the dipole-dipole interaction, in which an exchange magnon decays into two acoustic magnons.

and acoustic magnons is of fundamental importance for obtaining the amplitudes of interaction of the spin waves and will be taken into account systematically.

On the basis of everything said above, the structure of the Hamiltonian (6) is described schematically by the expression

$$\hat{H}_m=\sum_{k\mu}\varepsilon_{k\mu}\xi_{k\mu}{}^+\xi_{k\mu}+\sum_{1,2,3,4}\{I_1(1,2,3,4)\hat{O}_1\hat{O}_2\hat{O}_3\hat{O}_4$$
$$+I_2(1,2,3,4)\hat{O}_1\hat{O}_2\hat{A}_3\hat{A}_4$$
$$+I_3(1,2,3,4)\hat{A}_1\hat{A}_2\hat{A}_3\hat{A}_4+R_1(1,2,3,4)\hat{O}_1\hat{A}_2\hat{A}_3\hat{A}_4$$
$$+R_2(1,2,3,4)\hat{A}_1\hat{O}_2\hat{O}_3\hat{O}_4\}, \quad (14)$$

where $\xi^+_{k\mu}$ and $\xi_{k\mu}$ are the creation and annihilation operators of magnons on the branch μ with wave vector $\mathbf{k}$; $\varepsilon_{k\mu}$ is the energy of the magnons; the operators $\hat{A}_\sigma$ and $\hat{O}_\sigma$ of the form $\xi_\sigma \pm \xi^+_\sigma$ conventionally correspond to acoustic and exchange magnons, and σ is the collective index $\mathbf{k}\mu$. The amplitudes I in Eq. (14) are proportional to the exchange integrals, and R contains the quantity $R(\mathbf{k})$ from Eq. (8) in the form of a factor.

We note an important feature of the second term in braces in Eq. (14). It contains either four creation operators or four annihilation operators or a pair of creation operators and a pair of annihilation operators. In addition, in the last case one operator of the pair is an exchange operator and the other is an acoustic operator. Thus this term, describing in the exchange approximation the interaction of the exchange- and acoustic-magnon subsystems, contains only the scattering of exchange magnons by acoustic magnons.

The exchange part of the Hamiltonian of magnetoelastic interactions makes the greatest contribution to the relaxation of exchange spin waves via interaction with the elastic subsystem. It has the form

$$\hat{H}_{mp}=\sum_{\mathbf{k}\rho\mathbf{q}}\left(\left[\frac{k^2}{2NM_0\Omega_\rho(\mathbf{k})}\right]^{1/2}(b_{\mathbf{k}\rho}+b^+_{-\mathbf{k}\rho})\right.$$
$$\times\left\{\sum_{\sigma=1}^{3}[\lambda_\sigma^{(e)}(\mathbf{e}^\rho\mathbf{f})\mathbf{L}_\sigma(\mathbf{q})\mathbf{L}_\sigma(-\mathbf{k}-\mathbf{q})]\right.$$
$$\left.\left.+\lambda_4^{(e)}(e_xf_y+e_yf_x)[\mathbf{L}_1(\mathbf{q})\mathbf{L}_3(-\mathbf{k}-\mathbf{q})+\mathbf{L}_3(\mathbf{q})\mathbf{L}_1(-\mathbf{k}-\mathbf{q})]\right\}\right). \quad (15)$$

Here $\Omega_\rho(\mathbf{k})$ is the frequency of a phonon with momentum $\mathbf{k}$ on the branch ρ; M_0 is the mass of a unit cell; $\mathbf{e}_\rho$ is the polarization vector of the phonon; $\mathbf{f}=\mathbf{k}/k$; $b^+_{\mathbf{k}\rho}$ and $b_{\mathbf{k}\rho}$ are phonon creation and annihilation operators; $\lambda^{(e)}$ are exchange magnetoelastic constants. We write the Hamiltonian of the elastic subsystem in the standard manner:

$$\hat{H}_p=\sum_{\mathbf{k}\rho}\Omega_\rho(\mathbf{k})b_{\mathbf{k}\rho}{}^+b_{\mathbf{k}\rho}.$$

Substituting Eqs. (8) and (10) into Eq. (15), it is obvious that the part of the Hamiltonian (15) that is quadratic in the quasiparticle creation and annihilation operators contains the small factor D/J, describing the noncollinearity of the magnetic structure. For this reason, in what follows we shall neglect the mixing of the magnon and phonon states. The part of the Hamiltonian (15), that describes the magnon-phonon interaction processes has the following structure:

$$\hat{H}_{mp}=\sum_{1,2,3}(\lambda_{\rm I}^{(e)}\hat{A}_1\hat{O}_2+\lambda_{\rm II}^{(e)}\hat{O}_1\hat{O}_2)(b_3{}^++b_3). \quad (16)$$

We neglected the relativistic part of the Hamiltonian of the magnetoelastic interactions, since its structure is the same as that presented in Eq. (16).

3. RELAXATION PROCESSES

The lifetime of the exchange spin waves, as follows from Eq. (14) and (15), is determined by two groups of processes. The first group consists of decay processes. They include the process (I) in which an exchange magnon decays into three acoustic magnons and the process (II) in which an exchange magnon decays into an acoustic magnon and a phonon. The second group contains the following processes: III—merging of exchange and acoustic magnons and formation of two acoustic magnons, IV—scattering of an exchange magnon by an acoustic magnon, V—scattering of an exchange magnon by an exchange magnon, and VI—merging of an exchange magnon and an acoustic magnon and formation of a phonon. Taking into account the magnetic dipole interactions gives small corrections to all the processes studied above and results in a three-magnon process VII—the decay of an exchange magnon into two acoustic magnons. The Hamiltonian (1), in principle, admits other relaxation processes also, but they are forbidden by the conservation laws.

The damping coefficients of the exchange spin waves are calculated in the standard manner.[12] The imaginary part of the mass operator was calculated in second order of perturbation theory in the four- and three-particle interactions.

We shall now analyze the contributions to the imaginary part of the mass operator.

I. Decay of an exchange magnon into three acoustic magnons. The spin-wave energies calculated on the basis of the Hamiltonian (4), neglecting $\hat{H}_1(\mathbf{k})$, and the corresponding transformation coefficients u, v are presented in Refs. 3 and 4. In calculating the decay amplitudes only the largest terms were retained, i.e., the terms containing exchange-enhanced coefficients, of order $(J/A)^{1/4}$, of all three acoustic magnons. In this approximation only one decay channel described by the conservation law

$$\varepsilon_{1256}^{\pm}(0)=\varepsilon_{3478}^{\pm}(\mathbf{k})+\varepsilon_{3478}^{\mp}(\mathbf{q})+\varepsilon_{3478}^{\pm}(-\mathbf{k}-\mathbf{q}) \tag{17}$$

is realized for each exchange magnon with $\mathbf{k}=0$. The corresponding amplitude, found taking into account the mixing of the states of exchange and acoustic magnons, has the form

$$\Psi_{\mathrm{I}}^{\pm}(\mathbf{k},\mathbf{q})=\frac{i}{\sigma sN}[R(\mathbf{k})-R(\mathbf{q})]$$
$$\times d_{L_1}^{O\pm}(0)d_{L_3}^{A\pm}(\mathbf{k})t_{L_3}^{A\mp}(\mathbf{q})d_{L_3}^{A\pm}(\mathbf{k}+\mathbf{q}), \tag{18}$$

where we have introduced the notation $\nu=1256^{\pm}=O\pm$ and $\nu=3478^{\pm}=A\pm$, $R(\mathbf{k})=4is\tilde{R}(\mathbf{k})$, $\tilde{R}(\mathbf{k})$ is defined in Eq. (8), $d=u-v$, and $t=u+v$.

Since the general form of the amplitudes is extremely complicated, here and below we present expressions taking into account the explicit form of the combinations of the t, d coefficients and the momentum conservation law; this makes it possible to see clearly the interactions that determine these amplitudes.

In the range of fields $A_j \ll H \ll H_{sf}$ (where $A_j=A_y$ and A_z is the anisotropy field) and wave vectors $\mathbf{k}$ such that $\mathbf{ka}<1$, the energies of the acoustic magnons can be written in the form

$$\varepsilon_{A\pm}(\mathbf{k},H)=E_0(k_0^2+\tilde{k}^2)^{1/2}\pm H. \tag{19}$$

As a result the conservation laws (17) assume the form

$$(k_0^2+\tilde{k}^2)^{1/2}+(k_0^2+\tilde{q}^2)^{1/2}+(k_0^2+|k+q|^2)^{1/2}=(E_2E_3)^{1/2}/E_0=2\beta. \tag{20}$$

In these formulas E_0 and E_3 are determined by antiferromagnetic exchange integrals between the layers of magnetic atoms lying in the x, y-plane and correspondingly the ferromagnetic intralayer exchange; $E_2=E_0+E_3$ (Ref. 1); $(E_2E_3)^{1/2}$ is the energy of the exchange magnon in the absence of a field;

$$\tilde{\mathbf{k}}=\left\{\left(\frac{E_3}{E_0}\right)^{1/2}\frac{k_xa_x}{4},\ \left(\frac{E_3}{E_0}\right)^{1/2}\frac{k_ya_y}{4},\ \frac{k_za_z}{4}\right\},$$
$$k_0^2=\frac{A_y+A_z}{2E_0}; \tag{21}$$

A_y and A_z describe the anisotropy of the orthorhombic antiferromagnet. In the above approximations the magnetic field does not appear in the conservation laws (17); in addition, the t, d coefficients also do not depend on it and in the case of acoustic magnons they are determined by the expressions

$$d_{L_3}^{A\pm}(\mathbf{k})=\pm t_{L_2}^{A\pm}(\mathbf{k})=[4(\tilde{k}^2+k_0^2)]^{-1/4},$$
$$t_{L_3}^{A\pm}(\mathbf{k})=\pm d_{L_2}^{A\pm}(\mathbf{k})=[(\tilde{k}^2+k_0^2)/4]^{1/4}. \tag{22}$$

The line width of the exchange mode, governed by the decay process, has the form

$$\gamma_{\mathrm{I}}^{\pm}=\pi\sum|\Psi_{\mathrm{I}}^{\pm}(\mathbf{k},\mathbf{q})|^2[1+n(\varepsilon_A^{\pm}(\mathbf{k}))+n(\varepsilon_A^{\mp}(\mathbf{q}))$$
$$+n(\varepsilon_A^{\pm}(\mathbf{k}+\mathbf{q}))]\delta[\varepsilon_0^{\pm}(0)-\varepsilon_A^{\pm}(\mathbf{k})-\varepsilon_A^{\mp}(\mathbf{q})-\varepsilon_A^{\pm}(\mathbf{k}+\mathbf{q})], \tag{23}$$

where $n(\varepsilon)$ are the occupation numbers of magnons with energy ε. The range of the vectors $\tilde{\mathbf{k}}$ and $\tilde{\mathbf{q}}$ is found by analyzing the conservation laws (20), and to within terms of order k_0, small compared with β, the range is determined by the following limits:

$$\beta-\tilde{k}\leqslant\tilde{q}\leqslant\beta,\quad 0\leqslant\tilde{k}\leqslant\beta.$$

The integration over $\tilde{\mathbf{k}}$ is performed in a spherical coordinate system whose axes are oriented along the symmetry axes of the crystal and over the wave vector $\mathbf{q}$ in the spherical system whose z' axis is oriented along the vector $\tilde{\mathbf{k}}$ and whose y' axis lies in the x, y-plane.

As a result, at low temperatures $T\ll\varepsilon_{A\pm}(0)$ we obtain from Eq. (23)

$$\gamma_{\mathrm{I}}^{\pm}=\frac{8}{5}\left(\frac{4}{\pi}\right)^3\left(\frac{E_2}{E_3}\right)^{1/2}\left(\frac{E_0}{E_3}\right)^4\frac{R_0^2}{E_0}\left[\frac{\beta^6}{9}+\left(\beta^2\frac{T}{E_0}\right)^2\mathrm{ch}\left(\frac{H}{T}\right)\exp\left(-\frac{E_0k_0}{T}\right)\right]. \tag{24}$$

Here $R_0=8s\mathscr{K}_{12}^{yz}$, and terms with higher powers of T/E_0 have likewise been dropped. As expected, the processes in which an exchange magnon decays into acoustic magnons with large wave vectors (first term in brackets) make the greatest contribution to the half-width of the line. At the same time the temperature dependence of this process is determined by decay with the participation of acoustic magnons whose wave vectors lie near the center of the Brillouin zone (second term in brackets).

In the case of temperatures of the order of the activation energies of the acoustic magnons (i.e., for $T\sim\varepsilon^{\pm}(0)$) Eq. (23) can be represented in the form

$$\gamma_{\mathrm{I}}^{\pm}=\frac{8}{5}C\left\{\frac{\beta^6}{9}+\frac{T}{2E_0}\left[\beta^4(\varphi_1^{\pm}+\varphi_1^{\mp})-2\beta^3(\varphi_2^{\pm}+\varphi_2^{\mp})+2\beta^2\left(\frac{8}{3}\varphi_3^{\pm}+\varphi_3^{\mp}\right)-\beta\left(\frac{13}{3}\varphi_4^{\pm}+\varphi_4^{\mp}\right)+\frac{13}{15}\varphi_5^{\pm}+\frac{8}{15}\varphi_5^{\mp}\right]\right\}. \tag{25}$$

In this expression

$$C=\left(\frac{4}{\pi}\right)^3\left(\frac{E_2}{E_3}\right)^{1/2}\left(\frac{E_0}{E_3}\right)^4\frac{R_0^2}{E_0},$$
$$\varphi_m^{\pm}=\left(\frac{T}{E_0}\right)^m\int_{k_0E_0/T}^{\beta E_0/T}\frac{x^m\,dx}{\exp(x\pm H/T)-1}. \tag{26}$$

At temperatures $k_0E_0\ll T<E_0$ the terms in the brackets in Eq. (25) do not depend on the temperature and therefore the quantity γ_{I} increases linearly with the temperature.

II. The process in which an exchange magnon decays into an acoustic phonon and a magnon is described by the second term in the Hamiltonian (15). This term is of exchange origin and has no analogs in the two-sublattice model of an antiferromagnet, since the vectors $\mathbf{L}_2$ and $\mathbf{L}_3$ vanish in this model. The appearance of invariants of the form $U_{xy}\mathbf{L}_2\mathbf{L}_3$ is characteristic of multisublattice magnets. As analysis shows, in the decay process there arise acoustic phonons whose energies in a magnetic field behave in the same manner as the energy of the decaying exchange magnon, i.e., it either increases or decreases. As regards acoustic phonons, all three types of quasiparticles arise in the decay process.

The amplitudes of the corresponding processes have the form

$$\Psi_{\rm II}^{\rho\pm}(\mathbf{k}) = i\left[\frac{2}{M_0 N\Omega_\rho(\mathbf{k})}\right]^{1/2} \lambda_4^{(e)}\,[e_x^{(\rho)}k_y + e_y^{(\rho)}k_x]\, t_{L_1}^{O\pm}(0)\, t_{L_3}^{A\pm}(\mathbf{k}). \tag{27}$$

In this case the t, d coefficients of the acoustic magnons are exchange-weakened. In the exchange approximation, as $\mathbf{k}\to 0$ the amplitude of this process vanishes in accordance with Adler's principle.

The contribution of decay accompanied by the formation of a longitudinal phonon with $T\sim\varepsilon_A^\pm(0)$ to the line width is determined by the expression

$$\gamma_{\rm II}^{\parallel\pm} = \frac{1}{\pi}\frac{(\lambda_4^{(e)})^2}{E_2}\frac{k_B\Theta_D^\parallel}{\rho v_0 c_\parallel^2}\alpha_\parallel^5 \times\left\{1+\exp\left[-\frac{E_0}{2T}\left(\alpha_\parallel + \frac{\varepsilon_A^\pm(0)}{E_0}\right)\right]\right\}. \tag{28}$$

The total contribution to the line width in the case of formation of transverse phonons with two types of polarizations is equal to

$$\gamma_{\rm II}^{\perp\pm} = \frac{3}{5\pi}\frac{(\lambda_4^{(e)})^2}{E_2}\frac{k_B\Theta_D^\perp}{\rho v_0 c_\perp^2}\alpha_\perp^5 \times\left\{1+\exp\left[-\frac{E_0}{2T}\left(\alpha_\perp + \frac{\varepsilon_A^\pm(0)}{E_0}\right)\right]\right\}. \tag{29}$$

Here c_l is the velocity of sound on the branch l, ρ is the density of the crystal, v_0 is the cell volume, and the Debye temperature $\Theta_D^l = \hbar c_l/k_B v_0^{1/3}$; and,

$$\alpha_l = (E_2E_3)^{1/2}\cdot[\Theta_D^l + (E_0E_2/8)^{1/2}]^{-1}.$$

Analysis of the conservation law shows that the quantity α_l determines the characteristic value of the quasimomenta of the phonon and magnon which form as a result of decay. Since $\Theta_D^\parallel > \Theta_D^\perp$, the contribution of $\gamma_{\rm II}^\parallel$ is much smaller than that of $\gamma_{\rm II}^\perp$.

The first term in Eq. (15), which corresponds to the standard exchange magnetostriction, which also remains in the two-sublattice model, results in processes in which an exchange magnon decays into a phonon and an exchange magnon. Investigation of the conservation laws shows that in this specific case $\Theta_D > E_0, E_2$ this process is impossible. However, in magnets with the same magnetic structure, but with $\Theta_D \ll E_0, E_2$ these processes can contribute to the relaxation of the exchange modes.

III. Processes in which exchange and acoustic magnons merge and two acoustic magnons are formed. The amplitudes of these processes are proportional to the nonuniform Dzyaloshinskiĭ interaction. Taking into account the mixing of the states of the exchange and acoustic magnons as well as the direct contribution of $\hat{H}_1(\mathbf{k})$, in calculating the amplitudes we shall retain only the terms containing the exchange-enhanced t, d constants of all three acoustic magnons. The exchange magnon of this branch can merge with the acoustic magnons of both types, and the types of acoustic magnons arising in the process are determined by the conservation laws

$$\varepsilon_O^+(0)+\varepsilon_A^\pm(\mathbf{k}) = \varepsilon_A^+(\mathbf{q})+\varepsilon_A^\pm(\mathbf{k}+\mathbf{q}). \tag{30}$$

For O^- the signs in Eq. (30) must be inverted. A characteristic feature of all processes studied above is that by virtue of the phase relations between the coefficients t and d only processes whose conservation laws do not involve the magnetic field are allowed.

The amplitude of the merging processes have the form

$$\Psi_{\rm III}^{\pm(1)}(\mathbf{k},\mathbf{q}) = \frac{i}{\sigma s N}[R(\mathbf{k})-R(\mathbf{q})] \times t_F^{O\pm}(0)\,t_{L_2}^{A\pm}(\mathbf{k})\,t_{L_2}^{A\pm}(\mathbf{q})\,t_{L_2}^{A\pm}(\mathbf{k}+\mathbf{q}), \tag{31}$$

$$\Psi_{\rm III}^{\pm(2)}(\mathbf{k},\mathbf{q}) = i\frac{2R(\mathbf{q})-R(\mathbf{k})-R(\mathbf{k}-\mathbf{q})}{2\sigma N s} \times t_F^{O\pm}(0)\,t_{L_2}^{A\mp}(\mathbf{k})\,t_{L_2}^{A\pm}(\mathbf{q})\,t_{L_2}^{A\mp}(\mathbf{k}+\mathbf{q}). \tag{32}$$

The expressions (31) and (32) describe processes determined by the conservation laws (30) with the upper and lower sign, respectively.

For the contributions of these processes to the line width at low temperatures $T\ll\varepsilon_A^\pm(0, H)$ we obtain

$$\gamma_{\rm III}^{\pm(1)} = \frac{1}{30}C\beta^4 k_0\frac{T}{E_0}\exp\left[-\frac{\varepsilon_A^\pm(0,H)}{T}\right], \tag{33}$$

$$\gamma_{\rm III}^{\pm(2)} = \frac{1}{2}Ck_0^3\frac{T}{E_0}\exp\left[-\frac{\varepsilon_A^\mp(0,H)}{T}\right]. \tag{34}$$

As expected, the quantities $\gamma_{\rm III}^\pm$ in this case are determined by merging processes with long-wavelength thermal acoustic magnons. For this reason, aside from the fact that the quantities $\gamma_{\rm III}^\pm$ are exponentially smaller than $\gamma_{\rm I}^\pm$, Eqs. (33) and (34) contain the additional small parameter $k_0 \ll \beta$.

In the temperature range $T\gg\varepsilon_A^\pm(0, H)$ acoustic magnons with large wave vectors contribute to $\gamma_{\rm III}^\pm$:

$$\gamma_{\rm III}^{\pm(1)} = \frac{1}{30}\frac{CT}{E_0}\left(\beta^4\tilde\varphi_1^\pm + 2\beta^3\tilde\varphi_2^\pm - 2\beta^2\tilde\varphi_3^\pm - 3\beta\tilde\varphi_4^\pm + \frac{8}{15}\tilde\varphi_5^\pm\right), \tag{35}$$

$$\gamma_{\rm III}^{\pm(2)} = \frac{1}{2}\frac{CT}{E_0}\left(\beta^2\tilde\varphi_3^\mp + \beta\tilde\varphi_4^\mp - \frac{13}{22}\tilde\varphi_5^\mp\right). \tag{36}$$

Here the quantity $\tilde\varphi$ differs from the quantity determined in Eq. (26): in the latter quantity β is replaced by $k_{ZB}-\beta$, where $k_{ZB} = (\pi/4)(6E_3/\pi E_0)^{1/3}$ is the "effective" reciprocal lattice vector introduced with the help of Eq. (21).

IV. Processes in which exchange magnons are scattered by acoustic magnons. The amplitudes of these processes are of exchange origin, and the possible scattering channels are described by the conservation laws:

$$\varepsilon_O^+(0)+\varepsilon_A^\pm(\mathbf{k}) = \varepsilon_0^+(\mathbf{q})+\varepsilon_A^\pm(\mathbf{k}-\mathbf{q}). \tag{37}$$

The amplitudes of both scattering channels are equal to one another:

$$\Psi_{IV}^{(1,2)\pm} = -2\frac{E_0}{\sigma N s} t_{L_1}^{O+}(0)\, t_{L_3}^{A\pm}(\mathbf{k})\, t_{L_1}^{O+}(\mathbf{q})\, t_{L_3}^{A\pm}(\mathbf{k}+\mathbf{q}), \quad (38)$$

where the indices 1 and 2 refer to the upper and lower scattering channels (37), respectively. For the O^- magnons the signs in Eqs. (37) and (38) must be inverted. As follows from the definition (22), the t, d coefficients of both acoustic magnons are exchange-decreased. This is in agreement with Adler's principle, according to which the amplitudes of processes in which nonactivating quasiparticles participate vanish as the quasimomentum of any of these particles approaches zero.[14] In our case, the particles are nonactivational in the absence of a magnetic field in to the exchange approximation $k_0 = 0$. We shall calculate the contributions to the line width neglecting the dispersion of the t, d coefficients of the exchange magnons.

At low temperatures the scattering by long-wavelength acoustic magnons make the main contribution:

$$\gamma_{IV}^{\pm} = \gamma_{IV}^{\pm(1)} + \gamma_{IV}^{\pm(2)} = 4\left(\frac{4}{\pi}\right)^3 \frac{E_0}{E_3E_2} k_0^5 T\left(k_0 + 6\frac{T}{E_0}\right)\operatorname{ch}\left(\frac{H}{T}\right). \quad (39)$$

At temperatures $T \sim \varepsilon_A^{\pm}(0, H)$ we have

$$\gamma_{IV}^{\pm} = 12\left(\frac{4}{\pi}\right)^3 \frac{E_0^2}{E_3E_2} T\left\{\frac{T}{E_0}(\bar\varphi_5^{+} + \bar\varphi_5^{-}) - \frac{1}{3}\left(\frac{k_{ZB}}{2}\right)^6 \times \exp\left(-\frac{E_0 k_{ZB}}{2T}\right)\operatorname{ch}\left(\frac{H}{T}\right)\right\}. \quad (40)$$

Here $\bar\varphi^{\pm}$ is determined in Eq. (26), in which it is necessary to make the substitution $\beta \to k_{ZB}/2$. Thus, as one can see from Eqs. (39) and (40), in spite of the fact that the starting amplitude (38) is of exchange origin, the contribution of scattering of exchange magnons by acoustic magnons to the relaxation is small.

V. The processes corresponding to interactions within the exchange-magnon subsystem—these are processes in which an exchange magnon is scattered by another exchange magnon. It is obvious that in this case the number of thermal exchange magnons on which the scattering occurs is exponentially small at all temperatures right up to T_N. However, because of the weak dispersion of exchange spin waves, it should be expected that the region of k-space where scattering processes are allowed will be quite large. In addition, the amplitudes of these processes are proportional to the exchange and include the t, d coefficients, which are of order unity, of the exchange magnons. The scattering channels are determined by the conservation laws

$$\varepsilon_O^{+}(0) + \varepsilon_O^{\pm}(\mathbf{k}) = \varepsilon_O^{+}(\mathbf{q}) + \varepsilon_O^{\pm}(\mathbf{k}-\mathbf{q}), \quad (41)$$

where in the case of the scattering of an O^- magnon the signs must be inverted. The amplitudes of these processes, obtained neglecting dispersion of the t, d coefficients of the exchange magnons, have the form

$$\Psi_V^{(1)} = -\frac{2E_3}{\sigma s N}, \qquad \Psi_V^{(2)} = -\frac{E_0^2}{\sigma s N E_2}. \quad (42)$$

The number 1 in Eq. (42) corresponds to the process described by the expression (41) with the "+" sign and the number 2 corresponds to the process with the "−" sign. The contributions of these processes to the line width are determined by the expressions

$$\gamma_V^{(1)} = \left(\frac{E_3}{2E_0}\right)^2\left(\frac{\delta}{k_{ZB}}\right)^4 (E_2E_3)^{1/2}\exp\left[-\frac{(E_2E_3)^{1/2}+H}{T}\right],$$

$$\gamma_V^{(2)} = \frac{1}{4}\left(\frac{E_0}{2E_2}\right)^2\left(\frac{\delta}{k_{ZB}}\right)^4 (E_2E_3)^{1/2}\exp\left[-\frac{(E_2E_3)^{1/2}-H}{T}\right], \quad (43)$$

where $\delta = (2E_0)^{-1}[(E_3 + E_2)E_3]^{1/2}$. Here, as in all preceding expressions for γ, the dependence of the contributions of the field to the AFMR line width is determined by the field dependence of the occupation numbers of the thermal magnons, and for this reason the magnetic field enters only in the exponential.

VI. Magnon-phonon merging processes. In these processes an exchange magnon can merge with an acoustic or exchange thermal magnon and a phonon is formed. The amplitudes of these processes, which are proportional to the exchange magnetostriction constants of the second and first parts of the expression (15), respectively, have the form

$$\Psi_{VI}^{(1)\pm}(\mathbf{k}) = i\left[\frac{2}{M_0 N \Omega_\rho(\mathbf{k})}\right]^{1/2} \lambda_4^{(e)}(e_x^{(\rho)}k_y + e_y^{(\rho)}k_x)\, t_{L_1}^{O\pm}(0)\, t_{L_3}^{A\mp}(\mathbf{k}), \quad (44)$$

$$\Psi_{VI}^{(2)\pm}(\mathbf{k}) = i\left(\frac{k^2}{2NM_0\Omega_\rho(\mathbf{k})}\right)^{1/2} \mathbf{e}^\rho\mathbf{k}\,[(\lambda_2^{(e)} - \lambda_1^{(e)})\, t_{L_1}^{O\pm}(0)\, t_{L_1}^{O\mp}(\mathbf{k}) + (\lambda_3^{(e)} - \lambda_1^{(e)})\, t_F^{O\pm}(0)\, t_F^{O\mp}(\mathbf{k})]. \quad (45)$$

In the case of merging with an acoustic magnon phonons from all three branches can arise. The amplitude of these processes (44) contains the exchange-weakened t, d coefficients of the acoustic magnon. The corresponding total contribution to the line width is equal

$$\gamma_{VI}^{(1)\pm} = \frac{1}{2\pi}\frac{(\lambda_4^{(e)})^2}{E_2}\frac{k_B}{\rho v_0}\left\{\frac{3}{5}\frac{\Theta_D^{\perp}}{c_\perp^2}\tilde\alpha_\perp^5 \exp\left[-\frac{E_0\tilde\alpha_\perp + \varepsilon_A^{\mp}(0)}{2T}\right] + \frac{\Theta_D^{\parallel}}{c_\parallel^2}\tilde\alpha_\parallel^5 \exp\left[-\frac{E_0\tilde\alpha_\parallel + \varepsilon_A^{\mp}(0)}{2T}\right]\right\}, \quad (46)$$

where $\tilde\alpha_l = (E_2E_3)^{1/2}[\Theta_D^l - (E_0E_2/8)^{1/2}]^{-1}$ is the minimum wave vector of the acoustic magnon for which the merging process is possible. In this situation the presence of exponential factors means that by virtue of the relation $\Theta_D^{\perp} > \Theta_D^{\parallel}$ the merging processes in which longitudinal phonons are formed make a larger contribution than processes in which transverse phonons are formed.

We note two characteristics of the merging process with an exchange magnon: first, this process is possible only in magnets, where the condition $\Theta_D > 2(E_2E_3)^{1/2}$—twice the activation energy of the exchange magnons—is satisfied and, second, as follows from Eq. (45), only longitudinal phonons arise in the process. The calculation of its contribution gives

$$\gamma_{VI}^{(2)\pm} = \frac{2^3}{\pi E_2}\left[\lambda_2^{(e)} - \lambda_1^{(e)} - (\lambda_3^{(e)} - \lambda_1^{(e)})\frac{E_2}{E_3}\right]^2 \frac{k_B\Theta_D^{\parallel}}{\rho v_0 c_\parallel^2} \times\left(\frac{E_3}{E_2}\right)^{1/2}\left[\frac{(E_3E_2)^{1/2}}{\Theta_D^{\parallel}}\right]^{1/2}\exp\left[-\frac{(E_3E_2)^{1/2}\mp H}{2T}\right]. \quad (47)$$

Since Eq. (45) does not contain the exchange-weakened t, d

coefficients, the quantity $\gamma_{VI}^{(2)}$ can be comparable to $\gamma_{VI}^{(1)}$, in spite of the fact that the number of thermal exchange magnons is significantly smaller than the number of acoustic magnons with wave vectors $\tilde{\alpha}_{\parallel}$.

VII. We noted above that, in spite of the presence of the Dzyaloshinskiĭ interaction and the magnetic field, three-particle magnon-magnon processes, do not arise in the collinear phase by virtue of the symmetry restrictions. In this connection it is of interest to study the nonanalytic part of the dipole-dipole interaction, since for an arbitrary orientation of the wave vector **k** such restrictions should not arise. The Hamiltonian of this interaction has the form

$$\hat{H}_{d-d}=\frac{2\pi}{v_0}(g\mu_B)^2\sum_{\mathbf{k}}\frac{(\mathbf{kF}(\mathbf{k}))^2}{k^2}. \quad (48)$$

After transforming in Eq. (48) to the Goldstein–Primakov operators according to the formulas (8)–(13) and then making the t, d transformation we obtain a Hamiltonian in which we retain only the three-particle terms. The structure of this Hamiltonian is determined by the expression

$$\hat{H}_{d-d}^{(3)}=\sum_{1,2,3}[\eta_{123}^{(1)}\hat{A}_1\hat{A}_2\hat{A}_3+\eta_{123}^{(2)}\hat{O}_1\hat{O}_2\hat{A}_3$$
$$+\eta_{123}^{(3)}\hat{O}_1\hat{A}_2\hat{A}_3+\eta_{123}^{(4)}\hat{O}_1\hat{O}_2\hat{O}_3]. \quad (49)$$

Analysis of the conservation laws shows that only the process in which an exchange magnon decays into two acoustic magnons, as described by the third term in Eq. (49), is allowed. This process is possible only when the Dzyaloshinskiĭ interaction (both the uniform interaction—the terms proportional to a in the matrix $\hat{p}$ with the transformation from **L** to **L**′ and the inhomogeneous interaction, governed by the mixing of the states of the acoustic and exchange magnons) is taken into account. Its amplitude has the form

$$\Psi_{VII}^{\pm}=\frac{4\pi g\mu_B}{v_0(2\sigma sN)^{1/2}}\sum_{\nu=\pm}t_{L_1}^{O\pm}(\mathbf{q})\,t_{L_2}^{A\pm}(\mathbf{k}$$
$$+\mathbf{q})\left[\frac{D}{E_3}\frac{k_z}{k^2}+i\frac{R(\mathbf{k})}{E_3}\frac{k_x}{k^2}\right][k_y t_{L_3}^{A\nu}(\mathbf{k})-ik_z d_{L_2}^{A\nu}(\mathbf{k})]. \quad (50)$$

As follows from Eq. (22), the exchange-enhanced and exchange-weakened t, d coefficients of the acoustic magnons appear in this amplitude. For this reason, when calculating the contribution of this process to the line width, according to Eq. (22), we can set $t_{L_3}^{A\nu}(\mathbf{k})t_{L_2}^{A\nu}(\mathbf{k})=1/2$. As a result we have

$$\gamma_{VII}^{\pm}=\frac{8}{15\pi}\left(\frac{4\pi g\mu_B}{v_0}\right)^2\frac{1}{(E_3E_2)^{1/2}}\left[\left(\frac{D}{E_3}\right)^2(\beta_\pm^{\,2}+\beta_\mp^{\,2})\right.$$
$$\left.+\frac{8}{21}\left(\frac{R_0}{E_3}\right)^2\left(\frac{E_2}{E_3}\right)^2(\beta_\pm^{\,6}+\beta_\mp^{\,6})\right], \quad (51)$$

where $\beta_{\pm}=[(E_2E_3)^{1/2}\pm H]/2E_0$. In the formula (51) the terms containing exponentially small factors, describing the temperature dependence, were dropped. Comparing the contributions of the decay processes I and VII, we can see that the expression for γ_{VII}, in contrast to γ_I, contains the small parameter $(4\pi M)^2\cdot[E_3(E_3E_2)^{1/2}]^{-1}$, where $M=g\mu_B s/v_0$ is the magnetization of the sublattice. The dipole-dipole interaction Hamiltonian $\hat{H}_{d-d}^{(4)}$ that is fourth-order in the magnon creation-annihilation operators contributes to the amplitudes of all the above processes governed by the magnon-magnon interactions (14). However the magnitudes of these contributions are small because $4\pi M$ is small compared with J and D, and for this reason we did not take them into account anywhere. Thus the dipole-dipole interaction does not give rise to any additional four-particle relaxation processes for an exchange magnon; only the three-particle decay process VII makes a small contribution compared with the other processes.

EXPERIMENT. DISCUSSION

The experiment was performed on a spectrometer whose measuring cell is analogous to that described in Ref. 1 and which is equipped with a videographic digital information recording and processing system. The wavelength of the microwave generator (a backward-wave tube) was determined with an accuracy of 0.2% from the calibration curve measured beforehand with the help of an interference wavemeter. The relative error with which the frequencies were determined in the absorption spectra studied was governed by the stability of the voltage on the delay system of the generator and was equal to 0.01%. The spectrum was scanned with a pulsed magnetic field with a duration of 2 ms. The error in determining the magnitude of the field was less than 1%.

In order to satisfy the conditions of excitation of the exchange modes in the collinear phase (the magnetic vector of the microwave radiation $\mathbf{h}\parallel\mathbf{H}$ is polarized parallel to the x axis, which is also the a axis of the crystal) a pulsed Helmholtz coil was employed. The microwave radiation propagated along the b axis of the sample and the orientation of **h** could vary in the ac plane. The sample studied was placed in the channel of the solenoid between two quasioptic waveguides made of crystalline quartz, which with the help of conical lens junctions were matched with the radiation channeling duct. The measuring cell of the spectrometer ensures that the polarization is maintained and that the losses in the working wavelength range are small. The intensity of the magnetic field generated by the pulsed solenoids was determined from the integrated signal from the sensing coil, which was recorded simultaneously with the absorption spectrum of interest.

The working temperature was reached by evacuating helium from the cryostat and was determined to within ±10% from the vapor pressure. The radiation detector consisted of an n-InSb crystal, cooled to a temperature 4.2 K. The signal of the recorded absorption spectrum was digitized and stored in the controlling IBM PC computer, where it was processed together with the corresponding magnetic-field signal pulse. The use of original software made it possible to perform digital filtering of the noise and graphical analysis of the recorded spectrograms.

The samples for the investigation were prepared from $CuCl_2\cdot 2H_2O$ single crystals, grown from a solution of copper chloride. In the experiments $2.5\times2.5\times3.5$ mm^3 samples, which were faceted along the crystallographic axes, were employed. The Néel temperature of the investigated samples of $CuCl_2\cdot 2H_2O$ was equal to 4.33 K. The samples were oriented in two mutually perpendicular planes when the maximum splitting of the frequencies of the exchange modes in fields $H\approx H_{sf}$ was reached. The field **H** was orient-

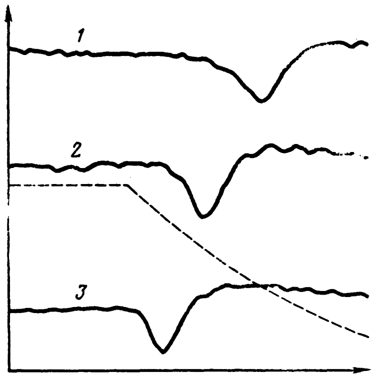

FIG. 2. Spectrograms of the absorption of the exchange mode in $CuCl_2 \cdot 2H_2O$ at the frequencies 8.98 cm^{-1} (*1*), 8.78 cm^{-1} (*2*), and 8.65 cm^{-1} (*3*) (the maximum of the magnetic field corresponds to 6.34 kOe); the dashed line shows the variation of the magnetic field.

ed with an accuracy of $\pm 3'$ parallel to the a axis in the ab plane, in which the sensitivity to the orientation was greatest. Although quite thick samples (3 mm) were employed, only 5–10% of the incident radiation was absorbed. In order to obtain spectrograms which are convenient to process the attenuation coefficient of the signal could be increased in a graduated fashion, so that the accuracy of the measurement of the intensity of absorption was always maintained.

Figure 2 shows a spectrogram of the absorption on the upper exchange mode ε_0^+ at three different frequencies at a temperature of 1.9 K. The half-width of the line is equal to $\gamma_0^+ = 750 \pm 75$ Oe. Since the energies of the exchange modes depend linearly on the field with $g = 2$, the line widths obtained in experiments with variable frequency and a constant field were the same as the line widths obtained with variable field and fixed frequency. At a constant temperature, as the measurements show, the line widths of both exchange modes are the same in the entire range of fields in which the collinear phase exists. Thus to within accuracy of the measurements the width of the absorption lines is independent of the strength of the field.

Figure 3 shows the change in the absorption line of the exchange mode ε_0^+ at the frequency 8.78 cm^{-1} in the temperature range 1.9–2.3 K. One can see that the line width increases as the temperature increases. However, we were not able to establish the specific character of the change in the line width, because the temperature of the sample was not measured accurately. The shift of the maximum of absorption of the exchange mode ε_0^+ in the region of higher fields as the temperature increases is governed by the general softening of the AFMR spectrum, which we studied previously in Ref. 20.

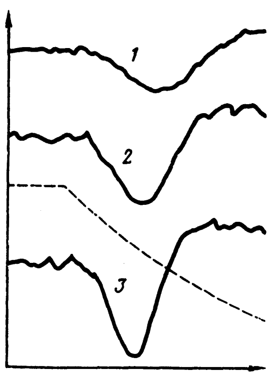

FIG. 3. The temperature dependence of the absorption line of the exchange mode in $CuCl_2 \cdot 2H_2O$ at the frequency 8.78 cm^{-1} (the maximum of the magnetic field corresponds to 5.67 kOe): $T = 2.2$ K (*1*), 2.0 K (*2*), and 1.9 K (*3*); the dashed line shows the variation of the magnetic field.

We now compare the theory and experiment. First we study the question of the intensity of absorption. The ratio of the intensity of the radiation ΔI absorbed by a crystal of thickness l to the intensity of the incident radiation I_0 is given by

$$\frac{\Delta I}{I_0} = \frac{8\pi s l (g\mu_B)^2}{\hbar c v_0} \left(\frac{D}{E_3}\right)^2 \left(\frac{E_3}{E_2}\right)^{1/2} \sum_{\nu=\pm} \frac{\gamma^\nu \omega^2 \varepsilon_0^\nu}{[\omega^2 - (\varepsilon_0^\nu)^2]^2 + 4(\omega\gamma^\nu)^2}. \quad (52)$$

This formula can be used to determine independently the strength of the Dzyaloshinskiĭ interaction from the known intensities of absorption at the maximum as well as the half-width and frequency of the resonance line.

In Ref. 1 we determined the values $E_3 = 36.5$ kOe, $E_0 = 150$ kOe, and $D = 2.5$ kOe. In Ref. 1 the quantity D was determined from an investigation of the region of "repulsion" of the branches of the frequency-field dependence of the exchange and acoustic modes in the spin-flop phase. According to the data of Ref. 17, we have $v_0 = 2.05 \cdot 10^{-22}$ cm^3. Substituting these values into the formula (52) and using the experimental value $\gamma = 0.75$ kOe, we find that at the maximum of absorption $\Delta I / I_0 \approx 0.015$ holds. This result is in good agreement with experiment.

In order to compare the theoretical and experimental values of the half-width of the line, we employ the results of Refs. 17–19 on the investigation of elastic and magnetoelastic properties of $CuCl_2 \cdot 2H_2O$. The parameters which we employ below are as follows: $\rho = 2.56$ g/cm^3, $c_\perp \approx 1.8 \cdot 10^5$ cm/s, and $c_\parallel \approx 3.4 \cdot 10^5$ cm/s (since the crystal is biaxial, here, of course, the values of the sound velocities are averages), $\Theta_D^\parallel \approx 45$ K and $\Theta_D^\perp \approx 23$ K. In Refs. 17 and 18 the exchange magnetoelastic constants $\lambda_1^{(e)} - \lambda_3^{(e)}$ were determined. Assuming that $\lambda_4^{(e)}$ is of the same order of magnitude, we obtain rough estimates $\lambda_4^{(e)} \approx \lambda_{1-3}^{(e)} \approx 12 \cdot 10^3$ kOe. As follows from Moriya's work,[15] $\mathcal{H}_{12}^{yz}$ can be of the same order of magnitude as $\mathcal{H}_{12}^{xz}$; in this case

$R_0 \approx D = 2.5$ kOe.

We shall estimate the contributions to the half-width of the line of the exchange mode on the basis of these values. At low temperatures $T \ll \varepsilon_A$ decay processes make the dominant contribution:

$\gamma_{I} \approx 4$ Oe, $\gamma_{II} \approx 200$ Oe.

At $T = 2$ K processes in which thermal quasiparticles participate make a measurable contribution:

$\gamma_{I} \approx 6$ Oe, $\gamma_{II} \approx 240$ Oe, $\gamma_{III} \approx 10$ Oe,

$\gamma_{IV} \approx 30$ Oe, $\gamma_{V} \approx 10$ Oe, $\gamma_{VI} \approx 15$ Oe.

Thus the total contribution of all processes to the half-width of the absorption line of the exchange modes is equal to $\gamma \approx 310$ Oe, which agrees satisfactorily with the experimentally determined value.

CONCLUSIONS

The above calculation of the line widths of the magnetic-resonance exchange modes in a four-sublattice orthorhombic antiferromagnet and a comparison of the results of this calculation with experimental data showed that the estimated total contribution to the damping of the exchange magnons agrees satisfactorily with the observed values. It seems that the two-particle scattering of exchange magnons by defects cannot make a significant contribution, because the dipole broadening of their spectra is significantly smaller than for acoustic magnons.

From the theoretical standpoint the weak dependence of the line width of the exchange magnons on the magnetic field in the collinear phase of the orthorhombic antiferromagnet is interesting and, at first, unexpected. This is because, on the one hand, the *t, d* coefficients of the exchange modes do not depend on the field owing to the linear field dependence of the energies of the exchange magnons in the collinear phase. On the other hand, the *t, d* coefficients of the acoustic magnons with large wave vectors (these are the acoustic magnons that participate in the elementary relaxation processes making the largest contribution) also do not depend on the field, since the spectrum of acoustic magnons also depends linearly on the field in this region of **k**-space. In addition, in the collinear phase of the orthorhombic antiferromagnet the Hamiltonian of the magnon-magnon interactions does not contain amplitudes that are proportional to the constant external field.

We call attention to the role of the Dzyaloshinskiĭ interaction, which determines both the intensity of absorption on exchange magnons (uniform Dzyaloshinskiĭ interaction)[6] and their line shape (the nonuniform Dzyaloshinskiĭ interaction is responsible for one of the main relaxation mechanisms).

As follows from our results, the relaxation of the exchange magnons is determined predominantly by decay processes. Decay processes governed by the magnetoelastic interaction of exchange origin make the largest contribution. The invariants in the magnetoelastic energy that lead to such processes can exist only in multisublattice magnets and are their characteristic feature. However the existence of these invariants is still not sufficient for the given exchange channel to be realized, since the channel is possible only if $2\Theta_D > \Omega_0$, where Ω_0 is the activation energy of the exchange magnon. For this reason it is of interest to investigate the line width of the exchange modes in objects with $2\Theta_D < \Omega_0$, where the lines apparently should be quite narrow.

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